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13. ABSTRACT (Maximum 200 words)
We summarize the studies done under this contract by listing the main accomplishments in the last 6 years. We started this research by fabricating arrays of antidots in a modulation doped FET structure using a focused ion beam technology. The unique transport properties in these antidot arrays were also studied. We extended the focused ion beam technology to the fabrication of zero dimensional resonant tunneling diodes and quantum wires fabricated using an insitu regrowth technique.

We then switched the research to the growth and studies of self assembled nanostructures. In the area of lateral superlattice growth and self assembling of quantum wire arrays, we have demonstrated MBE grown AlAs-GaAs lateral superlattice using transmission electron microscopy (TEM). We improved these self assembled lateral superlattices by growing the "serpentine superlattice"(SSL) using MBE. The SSL produces directly an array of quantum wires over a large wafer area. Using TEM, we were able to demonstrate AlAs-GaAs self assembled quantum wire arrays with adjustable dimensions. Optical studies of these self assembled quantum wires using polarizedphotoluminescence and photoluminescence excitation spectroscopy showed polarization effects that are associated with the 1D character of the structure. A quantum wire laser using the SSL growth method was then fabricated. As expected from the 1 D character of the structure, the quantum wire lasers shows large gain anisotropy at temperatures up to 150^{O}K. In a second phase of this contract we turned our efforts to the fabrication and studies of self assembled quantum dots . We first demonstrated a method for producing InAs-GaAs self assembled quantum dots (SAD) using MBE. These dots are ultra small (<20nm) and have a narrow size distribution. The zero dimensional character of these quantum dots using optical techniques was studied using optical techniques. Ultra narrow luminescence lines (FWHM 100 meV) have been demonstrated. The 0 D character of the structures persists at temperatures up to 150^{O}K and a strong room temperature luminescence is also observed at the important wavelength of 1.3 mm. We have then extended the self assembled quantum dots emission in the visible range using InAlAs-AlGaAs SAD grown by MBE. The 0D character of the SAD is also demonstrated by our observations of the sequential electron loading of the SAD using capacitance techniques. We demonstrate that these dots behave as artificial atoms at temperatures up to 110^{O}K. The ground state sequential electron loading of the SAD shows a Coulomb blockade effects at temperatures in excess of 100K has been observed in these SAD arrays.

14. SUBJECT TERMS
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FINAL REPORT

RESEARCH ON SELF ASSEMBLING NANOSTRUCTURES.

P.M.PETROFF, MATERIALS DEPARTMENT, UNIVERSITY OF CALIFORNIA, SANTA BARBARA. CA 93106

(AFOSR # F49620-92-J-0124)

Abstract:

We summarize the studies done under this contract by listing the main accomplishments in the last 6 years. We started this research by fabricating arrays of antidots in a modulation doped FET structure using a focused ion beam technology. The unique transport properties in these antidot arrays were also studied. We extended the focused ion beam technology to the fabrication of zero dimensional resonant tunneling diodes and quantum wires fabricated using an insitu regrowth technique.

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Optical studies of these self assembled quantum wires using polarized photoluminescence and photoluminescence excitation spectroscopy showed polarization effects that are associated with the 1D character of the structure. A quantum wire laser using the SSL growth method was then fabricated. As expected from the 1 D character of the structure, the quantum wire laser shows large gain anisotropy at temperatures up to 150^{o}K.

In a second phase of this contract we turned our efforts to the fabrication and studies of self assembled quantum dots . We first demonstrated a method for producing InAs-GaAs self assembled quantum dots (SAD) using MBE. These dots are ultra small (<20nm) and have a narrow size distribution. The zero dimensional character of these quantum dots using optical techniques was studied using optical techniques. Ultra narrow luminescence lines (FWHM 100 μeV) have been demonstrated. The 0 D character of the structures persists at temperatures up to 150^{o}K and a strong room temperature luminescence is also observed at the important wavelength of 1.3 μm. We have then extended the self assembled quantum dots emission in the visible range using InAlAs-AlGaAs SAD grown by MBE.

The 0D character of the SAD is also demonstrated by our observations of the sequential electron loading of the SAD using capacitance techniques. We demonstrate that these dots behave as artificial atoms at temperatures up to 110^{o}K. The ground state sequential electron loading of the SAD shows a Coulomb blockade effects at temperatures in excess of 100K has been observed in these SAD arrays.

STUDENTS AND POSTDOCTORAL RESEARCHERS working on the projects:
Z.Xu, J.J.Li, M.Miller, D. Leonard, G. Medeiros Ribeiro , and K.Pond.
Dr K.Schmidt and Dr. M.Oestreich, Dr.D.Mui, Dr S.Fafard, M.Wassermier, A.Lorke and Dr. R.Leon have also participated in research on these topics.

PhD degrees completed during the contract period: D.Leonard, M.Miller, Z.Xu, J.J.Li.

1) GROWTH OF QUANTUM WIRE SUPERLATTICES ON VICINAL SURFACES:

The research on the lateral superlattice has been aimed at characterizing the imperfections of these structures and looking for ways of improving their quality. A large effort was dedicated to the GaAs-AlGaAs serpentine superlattices (SSL). Both transmission electron microscopy (TEM) and photoluminescence (PL) and polarized PL spectroscopy measurements yielded important information on the main problem in these structures: the incomplete segregation between the GaAs and AlGaAs in the barrier and wire regions. The SSL structure in cross section TEM micrograph show the expected parabolic interfaces, however, the contrast between the well and barrier regions is weaker than expected for a perfect separation into AlAs and GaAs regions. The TEM micrograph, further points out the existence of an interface roughness.
A new and accurate method was developed for measuring both polarized photoluminescence (PL) and polarized photoluminescence excitation spectroscopy (PLE). The polarized PL measurements and their analysis indicate a segregation of 1/3 of the Al intended in the barrier into the wire region. A theoretical calculation indicates that the electron in their ground state clearly have a 1D character at the apex of the parabolas. However, theory and polarized PLE results indicate that the as grown GaAs-AlAs SSL structures confine only holes to 1D. Until a solution is found to remove the Al trapping into the wire regions, the electron will still have a 2D character in these structures.
The reason for the incomplete segregation have investigated by two methods. First, using scanning tunneling microscopy (STM), we have investigated the step edge roughness. The samples grown by MBE are transfered via a UHV suitcase into a separate UHV STM chamber where they are analyzed. For vicinal GaAs (001) surfaces, the step edge roughness is found to be commensurate with the surface reconstruction. Furthermore, we find that the step edge roughness could account in part for the poor Al segregation.

Second, using ultra short period $(AlAs)_1$- $(GaAs)_1$ superlattices we have started looking at the possible effects of Al-Ga surface exchange reactions during the formation of interfaces in the lateral superlattices. Our preliminary results, clearly indicate that exchange reactions between Al and Ga are important and could account for a large part of the intermixing observed by TEM and PL. We are continuing these investigations to try to solve the segregation problem.

2) GROWTH OF LATERAL SUPERLATTICES ON (110) GaAs VICINAL SURFACES:

To remove the intrinsic step edge roughness and possible that is due to the surface reconstruction of the (001) surface, we decided to investigate the quality of lateral superlattices deposited by MBE on vicinal (110) surfaces. The AlAs-GaAs system was studied because previous investigations indicated a strong tendency to a natural segregation between the AlAs and GaAs deposited by MBE on these surfaces. Experimental measurements using cross section TEM clearly indicate a better segregation of the AlAs and GaAs for these surfaces. However the vicinal GaAs(110) surface is strongly faceted for growth over a wide temperature range. We have modeled this faceting using a kinetic Monte Carlo method and compared the results of the time evolution of the facets with our model. A remarkable fit of the theory is obtained with experiments. Only one adjustable parameter, the fraction of deposited atoms going to the step riser of the terraces is used in the comparison between theory and experiments. Because of the inherent faceting problem due to the long diffusion length of Ga on the GaAs (110) vicinal surface, it seems that we should not continue growth studies of the lateral superlattice on this surface.

3) PHYSICAL PROPERTIES OF QUANTUM WIRE SUPERLATTICES:

We have recently started a series of transport measurements that in quantum wire structures (see View graph) with a short period (80Å) AlGaAs-GaAs grating.. These structures are doped by modulation doping of the quantum well cladding layer. The AlGaAs-GaAs is introduced by growing a thin (4 monolayers) lateral superlattice. A MODFET is fabricated and the gate voltage allows to sweep the Fermi level across the minigaps introduced by the grating. We expect that the structure will behave as a Bragg reflector. Preliminary results (4 growth runs) indicate that the magnetoconductivity versus gate voltage indeed indicate the presence of a plateau that is consistent with the existence of a minigap.

We have also carried out optical measurements on the quantum wire superlattices build in the serpentine superlattices in order to measure their carrier confinement properties. A unique photoelastic modulation technique was developed for simultaneously measuring the photoluminescence (PL) and the PL polarization in these samples. The results indicate a 2D confinement of the holes in the AlGaAs_GaAs quantum wire superlattices.
A quantum wire laser using the SSL growth method was then fabricated. As expected from the 1 D character of the structure, the quantum wire laser shows large gain anisotropy at temperatures up to 150^{o}K.

4) LATEST RESEARCH ACCOMPLISHMENTS:

For the period December 1994 to July 1995, a very large portion of our effort has been focused on the growth of self assembled quantum dots using controlled MBE deposition. Self ordering of the quantum dots has been demonstrated. A second major focus has been the spectroscopy of the self assembled quantum dots (SAQD).
The method is based on the strain induced nucleation of InAs dots grown on a GaAs substrate. The difference in lattice parameter (7%) is accommodated in two stages during growth. The first stage manifests itself by a transition from a 2 to 3 dimensional growth which gives rise to the InAs island that are coherently strained but dislocation free. The second stage of strain release is characterized by the introduction of misfit dislocations within the islands.
Here we use the self assembling coherently strained (dislocation free) islands to fabricate the quantum dots by embedding the InAs islands in GaAs. The processing does not require lithography or regrowth steps and is free of the carrier interface recombination processes that characterize most other processing approaches.
The existence of a critical thickness for the InAs wetting layer, allows us to develop a novel approach for the self positioning of the SAQD. We use the surface kinetics that are present at the intersection between two facets on a GaAs which was previously patterned to promote a local increase of the InAs wetting layer. Using the AFM, we can show the formation of a single line of islands with short range ordering between the islands. A computer simulation model which we have developed allows us to understand the interactions between the islands during the self ordering process.
We find that the quantum dot diameter can be adjusted between 10nm to 30nm. The size distribution around the mean value is better than ±5% and their density is adjustable from 10^{8} to 10^{10}cm^{-2} by proper choice of the growth parameter We have developed

SAQD in the InGaAs/GaAs system and the InAlAs/AlGaAs system. These show luminescence in the 1.1 micron and 0.68 micron range of the spectrum.

The photoluminescence (PL) and photoluminescence excitation (PLE) spectroscopy of the quantum dot samples show several characteristics that are consistent with three dimensional carrier confinement (ie. 0D structure).

Among these features we note:

a) The PL efficiency of the quantum dots is greater than 40 times that of a quantum well with the same thickness.

b) The PLE spectra show spectral peaks which can be interpreted as originating from the ground state and first excited state of the quantum dots.

c) The measured exciton binding energy in the quantum dots (12 meV) is greater than that of a quantum well (6meV).

d) The radiative lifetime in the quantum dot is temperature independent over a 40^{o}K temperature range .

e) The cathodoluminescence experiments and PL experiments probing very few quantum dots emitting in the visible range (620nm) extremely narrow emission lines. This is consistent with a delta function density of states for the quantum dots . We have continued our characterization of the SAQD electronic properties using magneto capacitance and FTIR measurements. The doping of the structure allows the loading of the quantum dots with electrons by applying a positive bias to the front gate. The magneto capacitance clearly shows sequential loading of the lower quantized subbands by electrons and holes. In addition, our magneto capacitance spectra show direct evidence of Coulomb charging (Coulomb blockade) for the electrons in the ground state of the SAQD. The Coulomb blockade is observed at temperatures in excess of 100K. This is a remarkable effect which is due to the small size of the SAQD and the large confining potential.

The FTIR spectra also show an absorption line corresponding to the excitation of electrons from the ground state to the first excited state energy level. The energy separation observed is the largest ever reported (40 meV) and corresponds well to that measured by resonant excitation PL measurements (32meV). In fact the magneto capacitance behavior of the loaded quantum dots is well accounted by a model that again assumes three dimensional carrier confinement.

In addition a splitting in the two dispersion branches of the magneto-absorption spectrum at zero magnetic field allows to detect an anisotropy in the SAQD shape. We find that the lateral confinement energies differ by a few meV because of the elliptical or rectangular shape of the SAQD.

In conclusion, the SAQD structures we have developed are the smallest and most efficient ones ever reported from a luminescence point of view. The quantum dot loading with electron has been observed a 4° and T>100°K for the first time and the lateral confinement energies (23 meV) are the largest ever reported.

5) DEVICE APPLICATIONS:

In view of the superior characteristics of these quantum dots, we have fabricated our first electrically injected quantum dot laser. The device operated at room temperature shows spontaneous emission at the wavelength of the quantum dots. However the lasing is observed to occur at a wavelength that is much higher in energies corresponding to that of the wetting layer that is connecting the dot. Obviously we have to work out a better laser structure to increase the filling factor by increasing the dot density. To better understand the SAQD laser properties, we have fabricated p-i-n structures with the SAQD layer in the i region of the device. Photoreflectance spectra, electroluminescence and photocurrent spectroscopy on these structures show the existence of excited states in the SAQD even at room temperature. This is a surprising result since thermal ionization of the carrier out of the SAQD is observed at T < 200K. Further studies of these devices are continued.

LIST OF PUBLICATIONS (AUGUST 1994-JULY 1995):

V.Bresller Hill, A.Lorke, K.Pond, P.M.Petroff and H.Weinberg. ***" The initial stages of InAs nucleation on GaAs"***Phys. Rev B 3, 2003 (1994).

Bressler-Hill, V.; Lorke, A.; Varma, S.; Petroff, P.M.; and others. ***" Initial stages of InAs epitaxy on vicinal GaAs(001)-(2*4)".*** Physical Review B , vol.50, (no.12):8479-87, (1994).

P.M. Petroff and S.P.DenBaars . ***" MBE and MOCVD growth and properties of self-assembling quantum dot arrays in III-V semiconductor structures."*** Superlattices and Microstructures, 1994, vol.15, (no.1):15-21.

D.S.Mui, D.Leonard, L.A.Coldren and P.M.Petroff ***"Surface migration induced self aligned Inas islands grown by molecular beam epitaxy"*** Appl. Phys. Lett. 66, 1620 (1995).

R.Leon, P.M.Petroff, D.Leonard and S.Fafard. ***"Spatially resolved visible luminesence of self assembled semiconductor quantum dots"*** Science, 267, 1966 (1995).

Fafard, S.; Leon, R.; Leonard, D.; Merz, J.L. and P.M.Petroff ***"Zero dimensional induced optical properties in self-assembled quantum dots. "*** J. of Superlattices and Microstructures, 1994, vol.16, (no.3):303-9.

V.Bressler-Hill, S.Varma, A.Lorke, B.Z. Nosho, P.M.Petroff and W.H.Weinberg ***"Island scaling in strained hetero epitaxy : InAs/GaAs (100)"*** Phys Rev Lett. 74, 3209 (1995).

G.Medeiros Ribeiro, D.Leonard and P.M.Petroff. ***" Electrons and holes energy levels in InAs self assembled quantum dots."*** Appl. Phys. Lett. 66, 1767 (1995).

R.Leon, S.Fafard, D.Leonard, J.L.Merz and P.M.Petroff ***" Visible luminescence from semiconductor quantum dots in large ensembles"*** Appl. Phys. Lett. 67, 521, (1995).

K.Eberl, P.M.Petroff and P.Demeester, ***"Low dimensional strucutres prepared by epitaxial growth or regrowth or patterned substrates"*** Book, NATO ASI Series (Kluver, Dortrecht, 1995).

DenBaars, S.P.; Reaves, C.M.; Bressler-Hill, V.; Varma, S. H.Weinberg and P.M.Petroff" ***Formation of coherently strained self-assembled InP quantum islands on InGaP/GaAs(001).*** Journal of Crystal Growth, Dec. 1994, vol.145, (no.1-4):721-7.

Leon, R.; Margolese, D.; Stucky, G.; Petroff, P.M. ***"Nanocrystalline Ge filaments in the pores of a mesosilicate."*** Physical Review B (Condensed Matter), 15 July 1995, vol.52, (no.4):R2285-8.

Fafard, S.; Leon, R.; Leonard, D.; Merz, J.L. and P.M.Petroff. ***"Phonons and radiative recombination in self-assembled quantum dots."*** Physical Review B (Condensed Matter), 15 Aug. 1995, vol.52, (no.8):5752-5.

PATENTS:

" Method for the fabrication of serpentine superlattices and quantum wire arrays" Patent pending.

" Method for the growth of self assembled quantum dots" Patent application submitted.

PROPERTIES OF QUANTUM WIRE LASERS. AFOSR (F4962J92-J-0124)

P.M.Petroff. University of California. Santa Barbara.

Method and principle:
@ Uses MBE deposition to directly form by self assembling epitaxy the quantum wires.
@ Uses the quantum wire serpentine superlattice (SSL) in the gain region to form a separate confinement heterostructure (SCH) in plane ridge waveguide device laser.

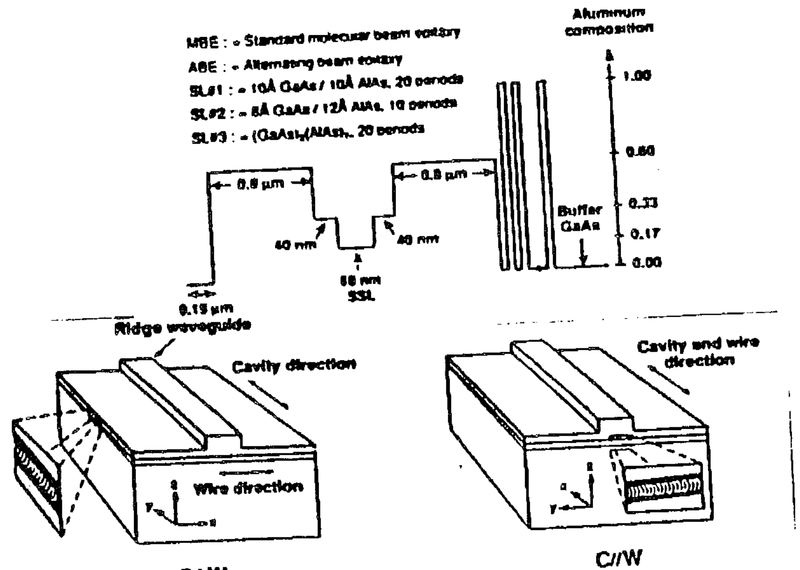


Figure 1: Band Diagram and laser structure.

@ Gain spectra are measured for ridge wave guide lasers parallel or perpendicular to the wire axis.

Results: The optical gain for the TM mode becomes greater than for the TE mode when the optical cavity is parallel to the nanowires axis. Theory modeling confirmed the 1D character of these devices.

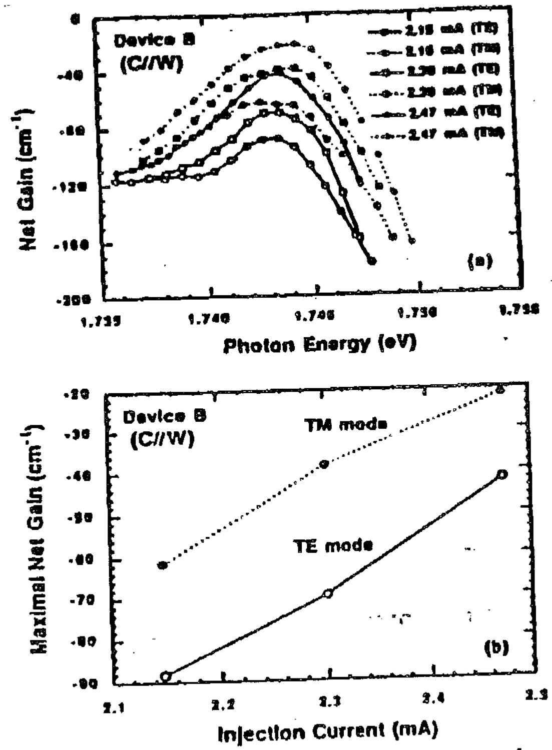


Figure2: Net gain spectra for both TE and TM mode measured for a device with wire axis parallel to the cavity .

@The 1D properties of this laser where observed to 100oK.

PROPERTIES OF QUANTUM DOTS LASERS. AFOSR (F4962J92-J-0124)

P.M.Petroff. University of California. Santa Barbara.

Method and Principle:

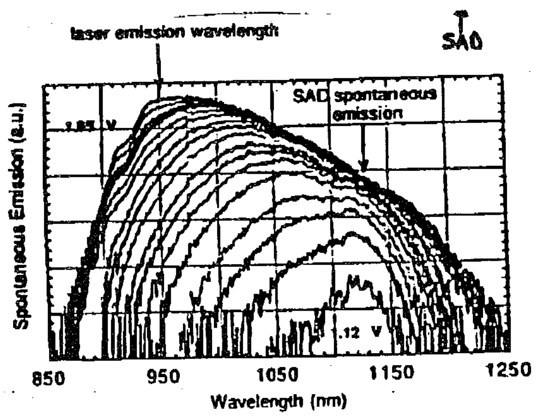
@Uses MBE deposition to directly form by self assembling growth the quantum dots.

@Uses the self assembling quantum dots in the gain region to form a separate confinement heterostructure in plane ridge waveguide laser.

@ The band diagram is similar to that of the quantum wire structure but the quantum wire layer is replaced by an InAs quantum dot layer.

@ Measurement of the electroluminescence spectrum at room temperature and CW.

@ Measurement of the L-I characteristics.



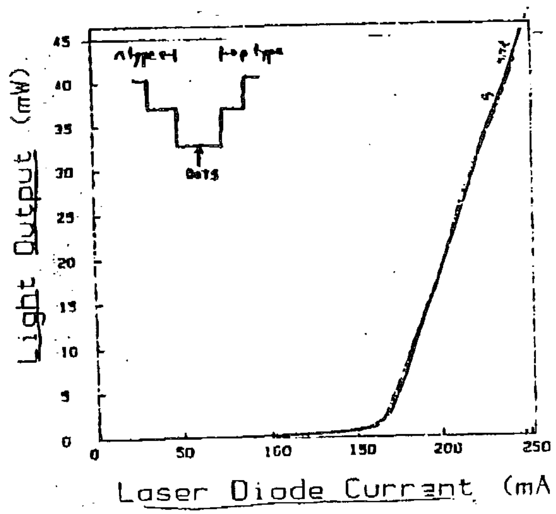


Figure 1: L-I Characteristics and electroluminescence spectra for a quantum dot laser. T=300K and CW operation.

RESULTS: The quantum dot laser lases at a higher energy than the expected value and we are attempting to understand this result and measure gain spectra to demonstrate that the 0 dimensional character of the laser.

GROWTH AND PROPERTIES OF SELF ASSEMBLED QUANTUM DOTS. AFOSR (# F 49620-92-J-0124)

P.M.PETROFF. University of California. Santa Barbara.

METHOD AND PRINCIPLE:

@ Uses MBE deposition of InAs or AlInAs on GaAs or AlGaAs.

@ Misfit strain produces a transition from 2D to 3 D growth for the In rich layer. This will produce islands on the surface. RHEED is used to detect the islands.

@ Capping of the InAlAs or InAs islands with a wider band gap material yields self assembled quantum dots (SAD).

RESULTS:

@ Ultra small (diameter <30nm,height 4nm)SAD hemispherical shaped islands are observed by TEM and AFM. The SAD formed by capping with a GaAs layer are defect free.

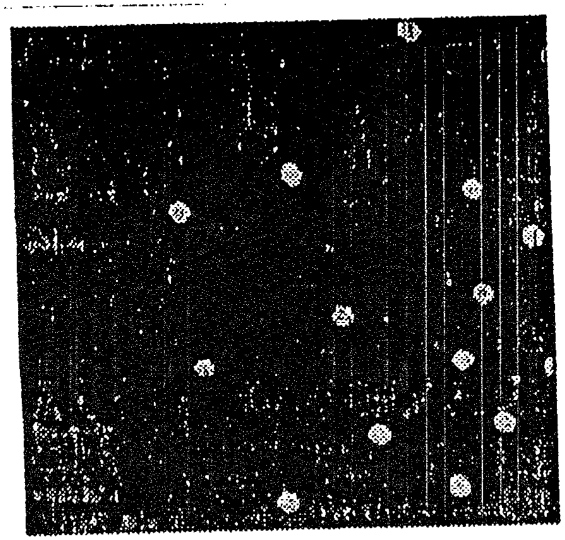


Figure 1: Atomic force micrograph of InAs self assembled islands.The islands are nucleating at step edges on the surface.

@ Very narrow size distribution (10-15% diameter, 1-2 monolayer height) is observed.

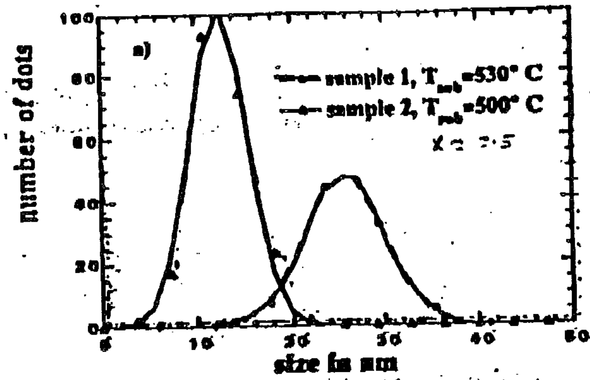


Figure 2: AFM measured diameter distribution of InAs islands for 2 different deposition temperatures.

@ A first order phase transition from 2D to 3D growth is observed. This allows for a large range of SAD densities (10^6 to $10^{10}cm^{-2}$) and the tightest size dispersion.

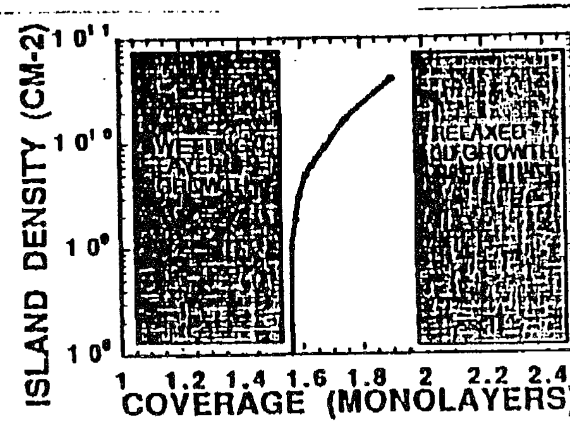


Figure 3: Measured density of InAs islands as a function of In flux on the wafer. The In coverage dependence of the island density is consistent with a first order phase transition.

OPTICAL PROPERTIES OF SELF ASSEMBLED QUANTUM DOTS.

P.M PETROFF, University of California, Santa Barbara.

AFOSR (Contract # F 49620-92-J-0124)

GOAL:
@Demonstration of the 0D character of the self assembled quantum dots.

METHOD:
@ Photoluminescence (PL) and photoluminescence excitation (PLE) spectroscopy are used.
@ Low temperature cathodoluminescence (CL) is used for imaging of a single dot.

RESULTS:
@ PL and PLE spectra of the SAD indicate a large energy shift (50-70meV). This is interpreted by assigning the first PLE peak to the first excited stated of the SAD.
The ground state excitation is not seen in the PLE spectra because it is an extremely narrow (<0.1meV) and weak line.

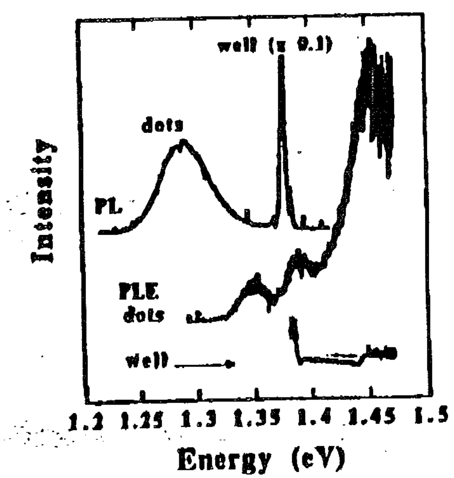


Figure 1: PL and PLE spectra of a sample containing an InAs SAD layer and a test quantum well.

@ As expected from a 0D system, the radiative lifetime of the SAD is independent of the temperature at temperature up to 40°K.

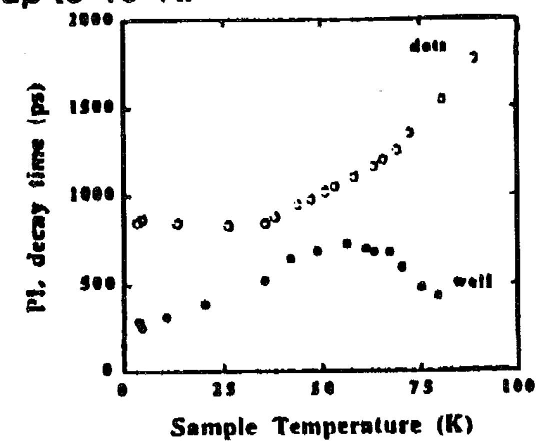


Figure 2: Radiative lifetime as a function of temperature for InAs SAD and an InGaAs quantum well.

@ Ultra narrow (0.4meV<) PL line width are observed from individual quantum dots.

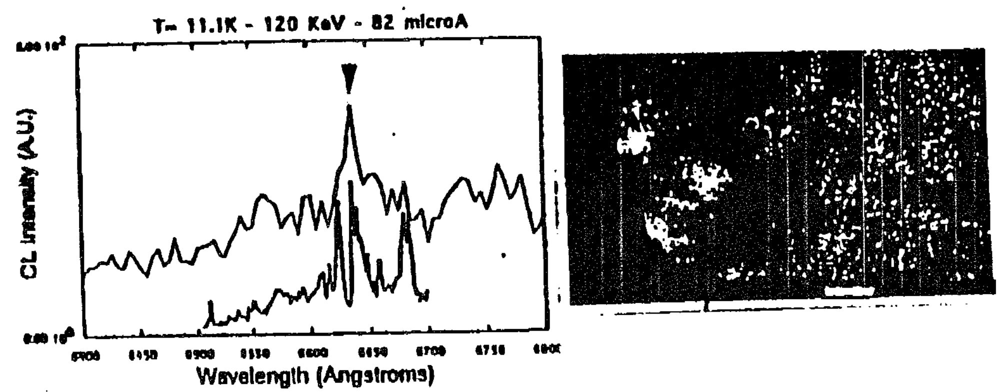


Figure 3: Cathodoluminescence spectrum and spectrally resolved images of individual quantum dots (Bright areas).

ELECTRONIC PROPERTIES OF InAs SELF ASSEMBLED QUANTUM DOTS.

P.M.PETROFF. University of California. Santa Barbara.

AFOSR (Contract # F 49620-92-J-0124)

GOAL:
@Demonstration of the electron and hole loading of the self assembled quantum dots.

METHOD:
@ Include the SAD into a MISFET device to detect the electronic loading of the self assembled dots. The measured device capacitance is proportional to the density of states in the dots.
@ Infrared absorption spectroscopy to detect the excitation of the electron from the ground state of the 1st and 2d electronic excited states.

RESULTS:

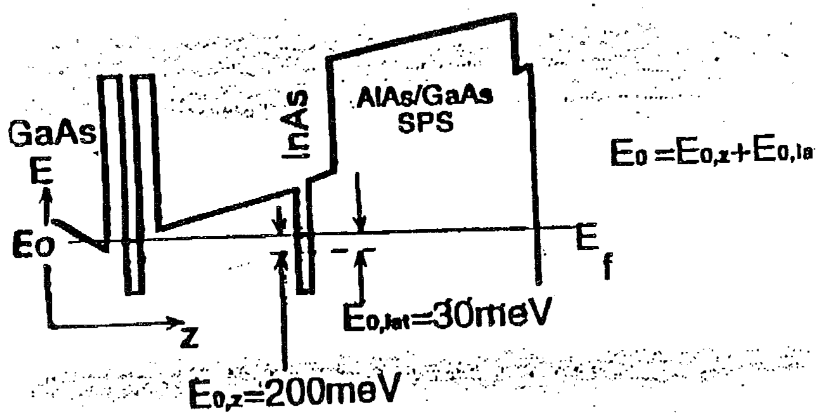


Figure 1: Schematic of the conduction band for the MISFET structure that includes the SADs. Positive bias of the top Shottky contact will bring the dot level in resonance with the Fermi level and allow electron tunneling into the dots. The changes in the capacitance are detected.

@ The CV characteristics indicates loading of and 2 electrons in the ground electronic state of the SAD. The third broad peak corresponds to the loading of electrons in the first excited state. The separation of the first 2 peaks corresponds to the Coulomb barrier seen by the 2d electron in the ground state. This Coulomb barrier is still seen at 77°K.

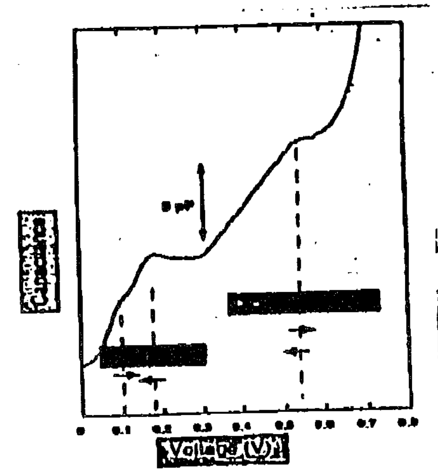


Figure 2: CV characteristic of an ensemble of ~10^6 InAs self assembled quantum dots.
@ In the IR absorption, the excitation of electrons between the 1st and 2d subband is seen at precisely the voltage which corresponds to the loading of the first electrons in the capacitance experiment.
@ The CV characteristics indicate a 0D density of state for a SAD ensemble. For the first time we have structures where **the Coulomb blockade energy (18-20meV) is larger than the intersubband energy (~40meV).**
@ Consequently we have been able to create artificial atoms.